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Mechanism for recombination of radiation-induced point defects at interphase boundaries

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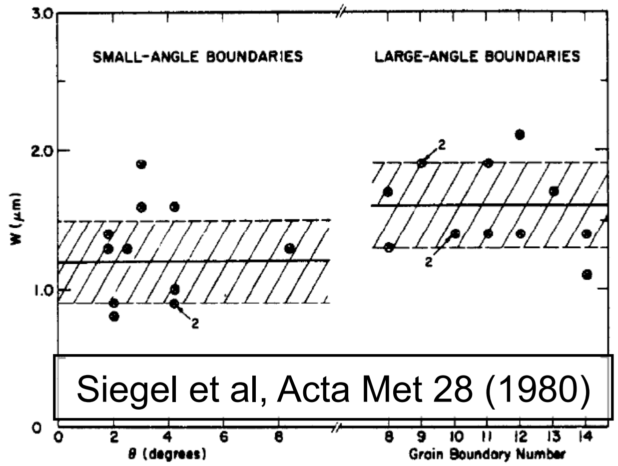
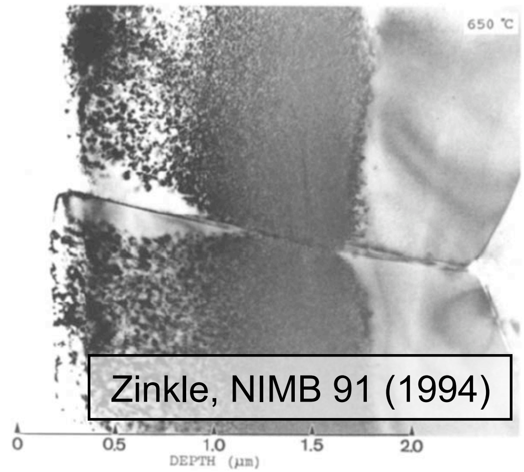
Boundaries and interfaces as sinks: Perspective/history

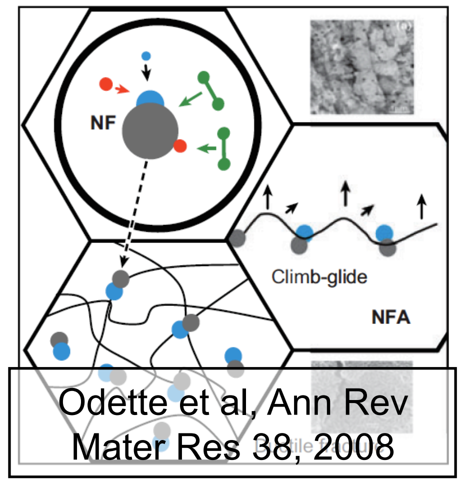
- Well established that grain boundaries (GBs)/interfaces are defect



Siegel et al, Acta Met 28 (1980)



Zinkle, NIMB 91 (1994)



Odette et al, Ann Rev Mater Res 38, 2008

- Atomistic details/time evolution of defects near boundaries still unclear

Earlier work and motivations of this work

- Earlier, Bai et al. proposed an interstitial emission mechanism (Bai *et al.*, Science 327, 1631 (2010)) for high-symmetry special GB in single-phase fcc Cu: interstitials observed to reemit from GB to recombine with vacancies near GB.
- Motivation from materials stability consideration:
  - GBs wold be subjected to rapid grain growth during irradiation and, therefore, the effectiveness of this mechanism may be lost.
  - For immiscible composites, the nanoscale length scales in the as-synthesized material may be retained during irradiation
- Motivation from "delocalization" question:
  - With a delocalized displacement field around an interstitial, the interstitial may effectively "lose its identity" in the interphase boundary
  - Does the interstitial emission mechanism still hold?

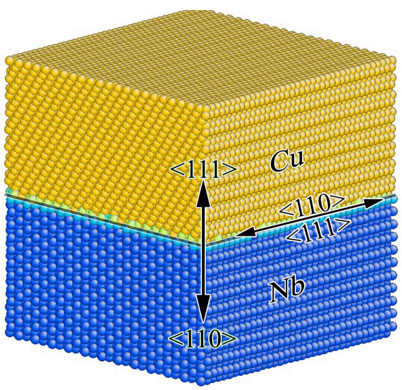
Heterogeneous Cu-Nb interphase boundaries

- Experimentally, nanolayered Cu-Nb exhibit significantly enhanced radiation-damage tolerance.
- Atomistic modeling studies of defect-interface interactions at fcc-bcc interfaces, Cu-Nb, as a model system.

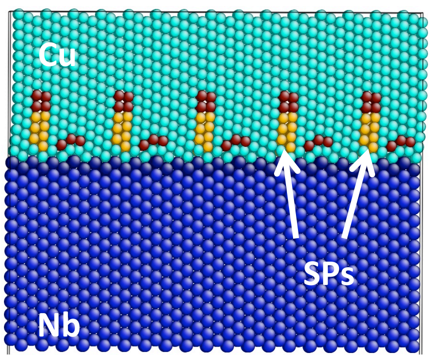
- Multilayer Cu-Nb interface Kurdjumov-Sachs (KS) orientation relation (OR):

  $\{111\}_{Cu}||\{110\}_{Nb}$ habit plane and $\langle 110\rangle_{Cu}||\langle 111\rangle_{Nb}$

- Severely plastic deformed (SPD) Cu-Nb interface with KS OR, but $\{112\}_{Cu}||\{112\}_{Nb}$ habit plane



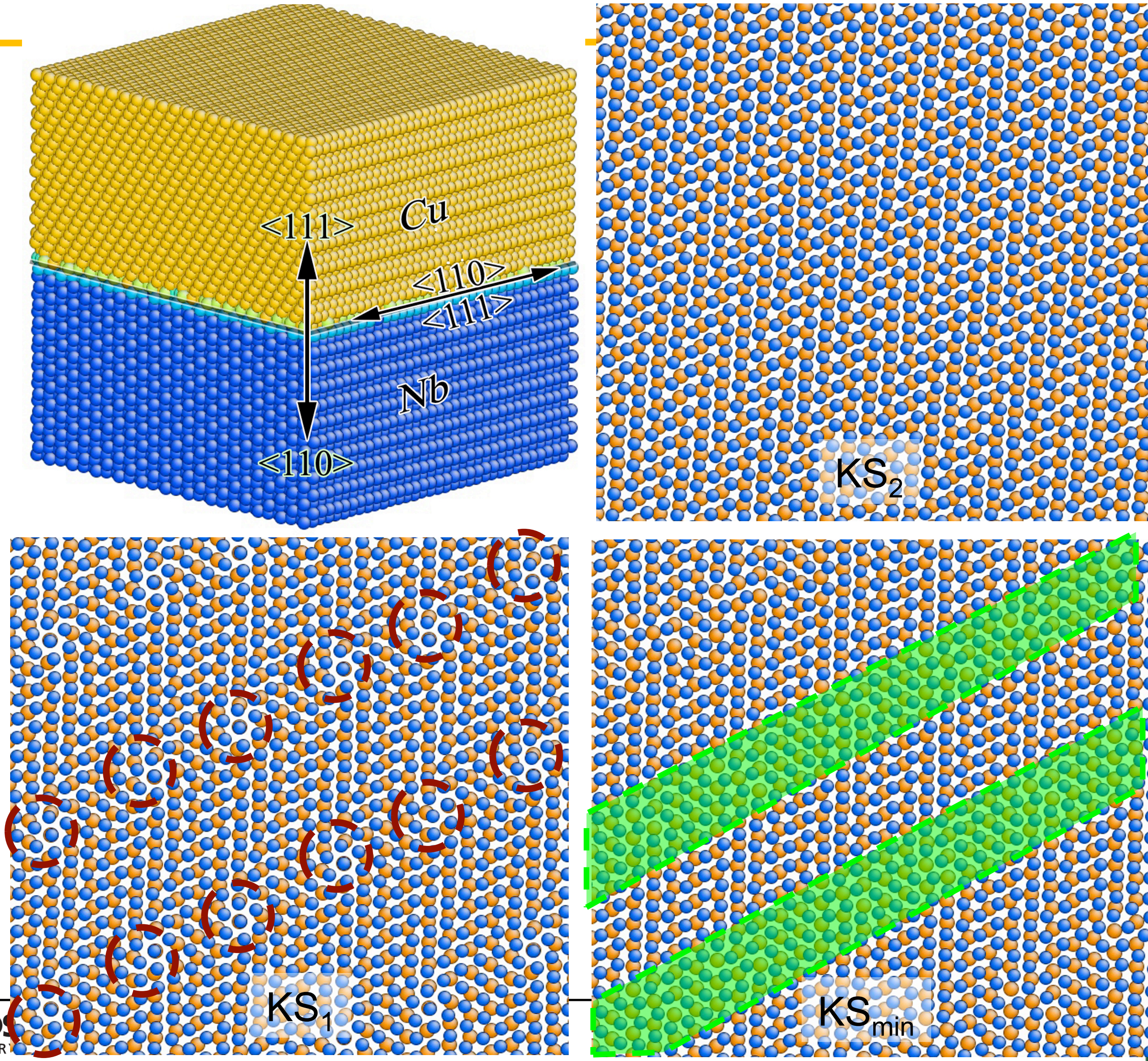
(KS interface)



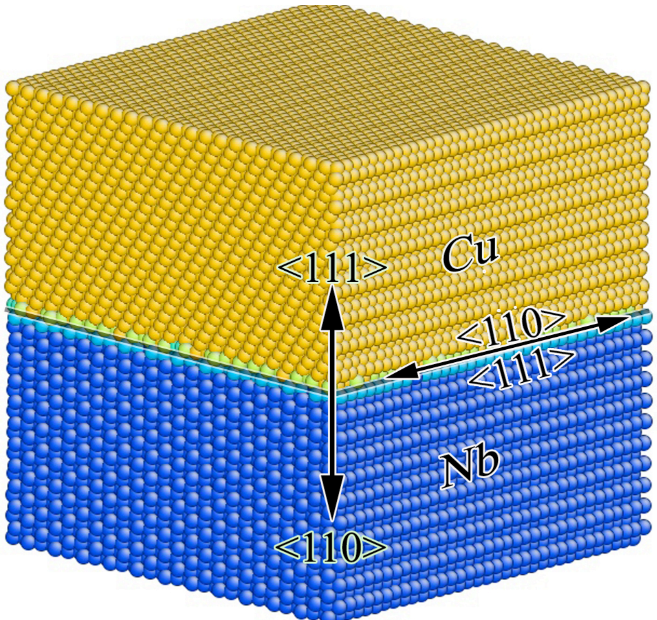
(SPD interface)

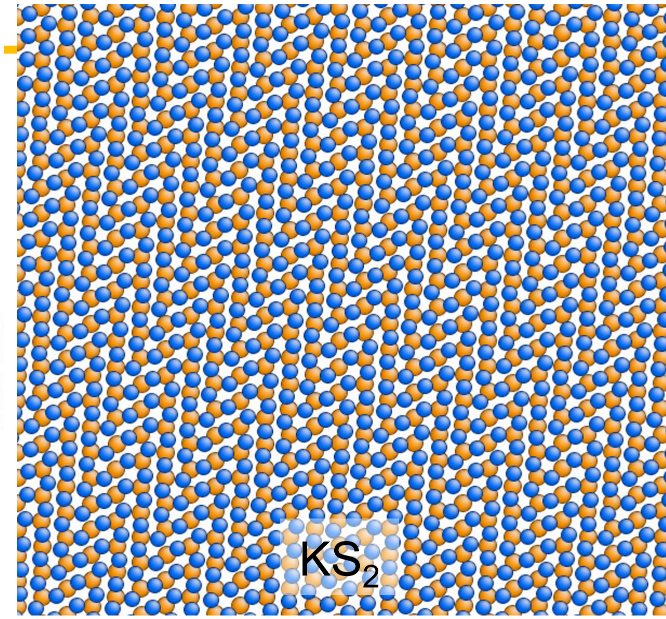
- Variation of KS interfaces using “tunable potentials”

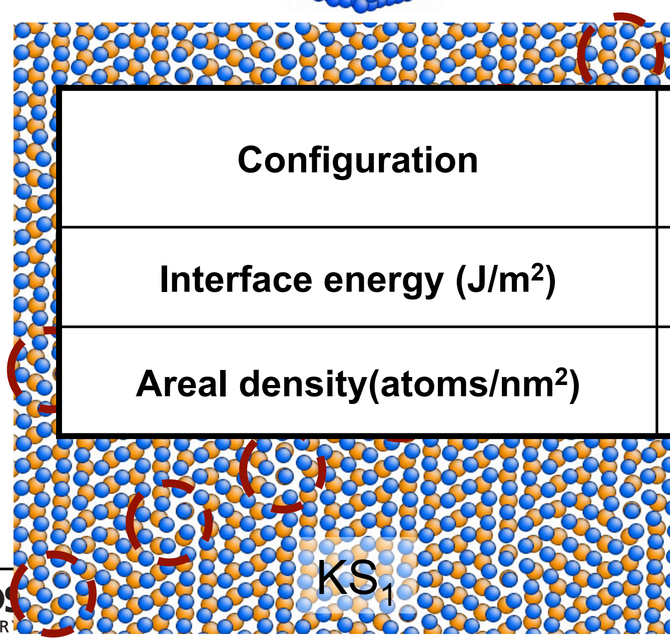
Atomistic modeling of fcc/bcc interfaces reveals multiple states of atomic structures with nearly degenerate formation energy

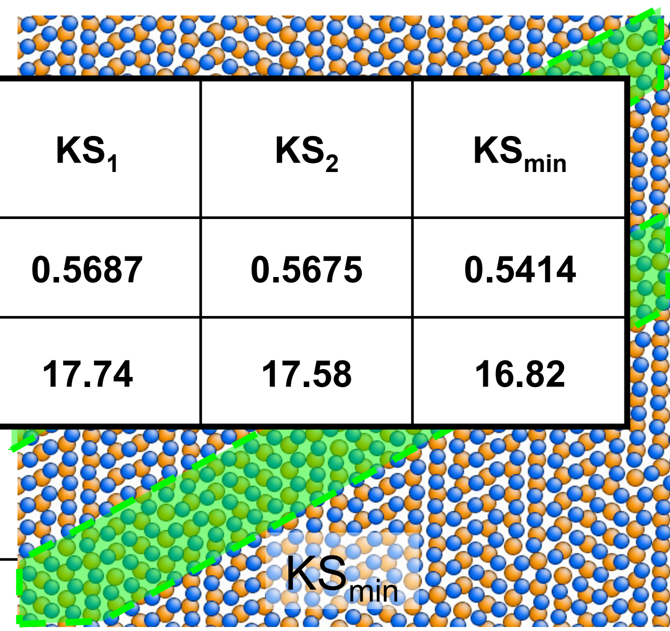


Atomistic modeling of fcc/bcc interfaces reveals multiple states of atomic structures with nearly degenerate formation energy



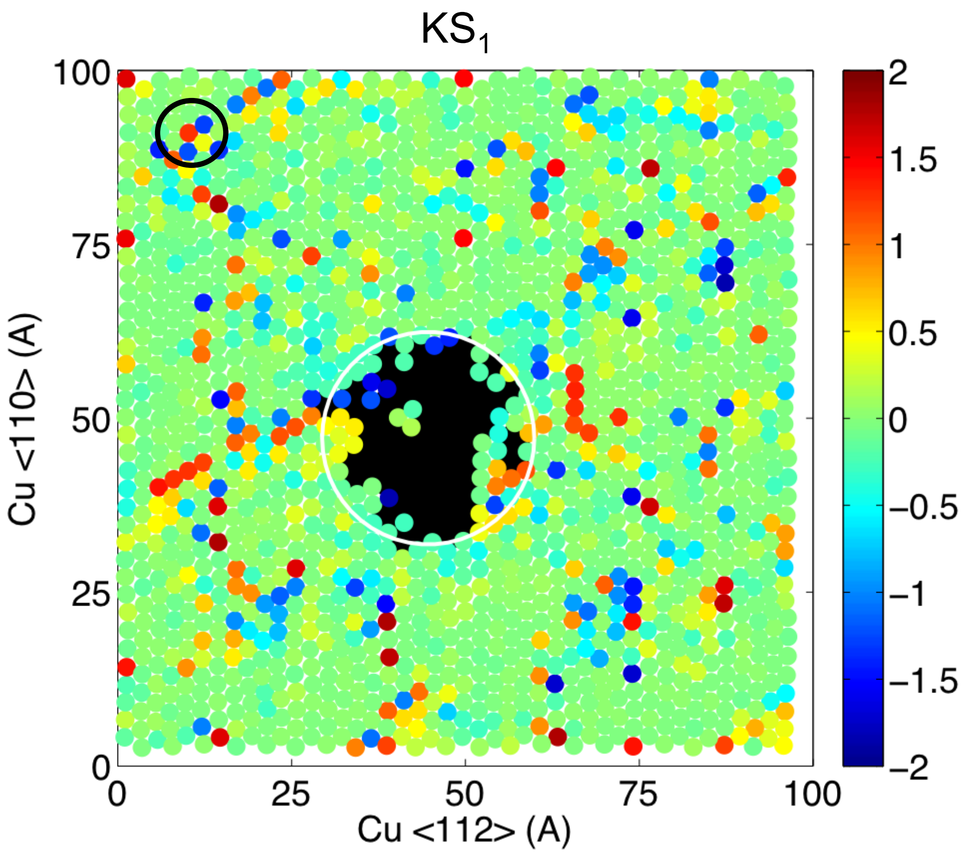




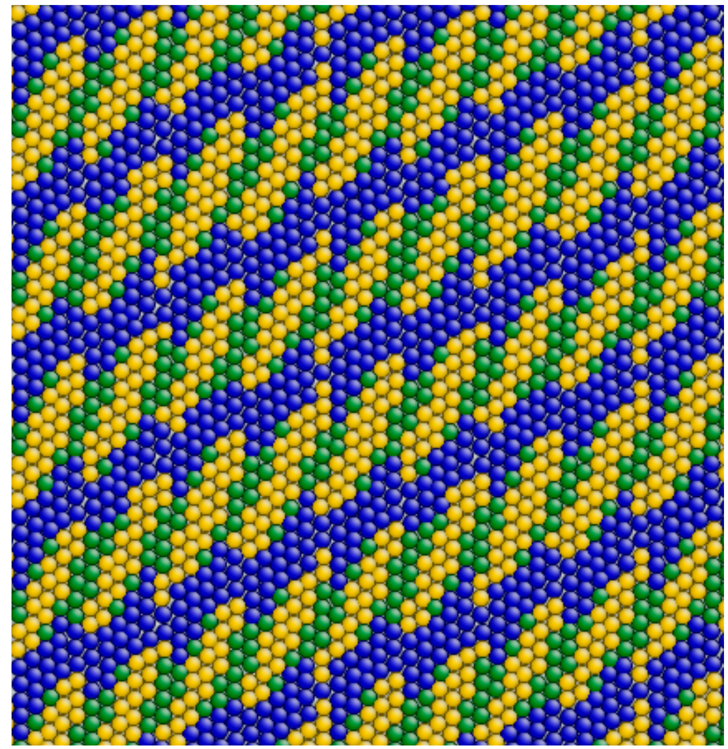


Configuration	KS_1	KS_2	KS_{min}
Interface energy (J/m^2)	0.5687	0.5675	0.5414
Areal density($atoms/nm^2$)	17.74	17.58	16.82

Delocalization at KS Cu-Nb interfaces



Within an interface, defects delocalize.

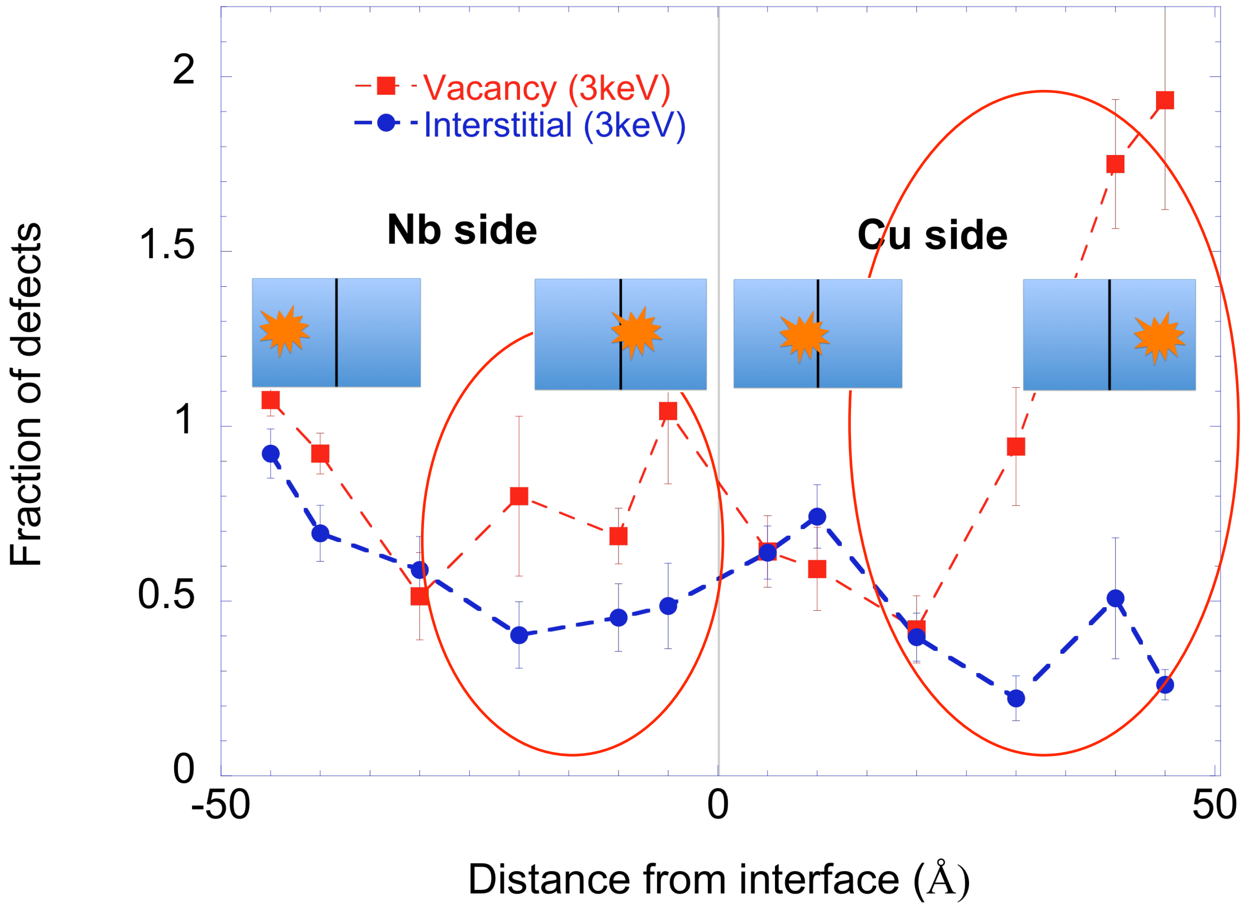


Dislocation network at interface.

In the SPD interface, point defects absorbed remain compact

Collision cascade MD simulations indicate asymmetric behavior across Cu-Nb interface

Radiation-induced point defects as a function of initial PKA position



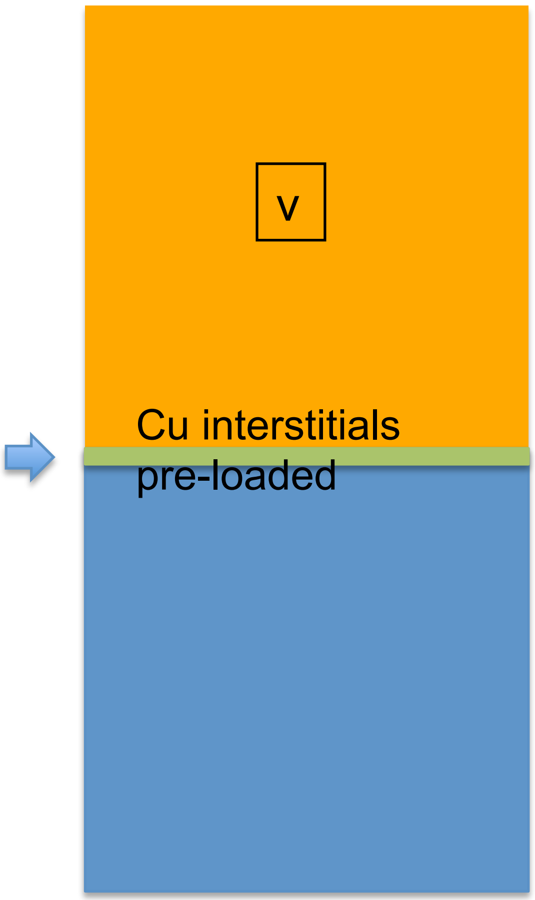
- KS_1 Cu-Nb interface
- PKA energy 3 keV

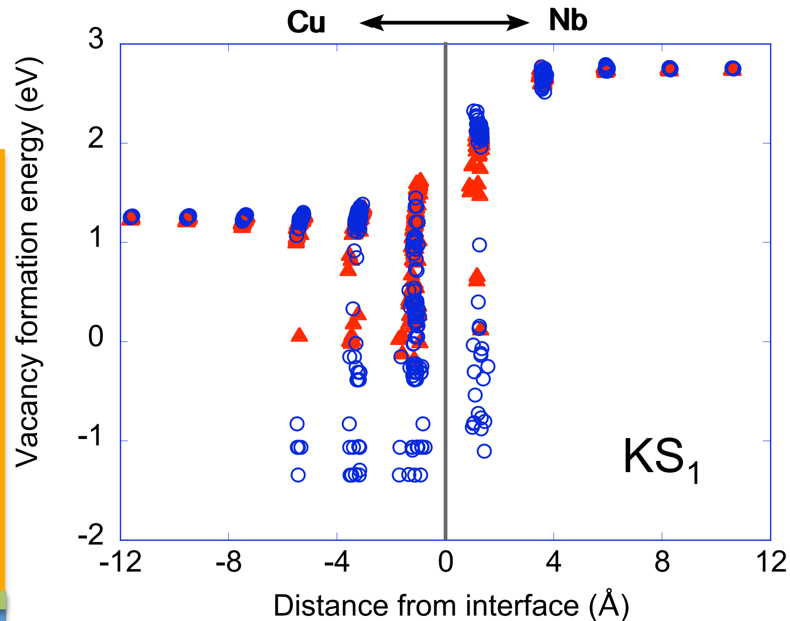
$$F_{defects} = \frac{n_{Cu}}{n_{Cu}^{bulk}} + \frac{n_{Nb}}{n_{Nb}^{bulk}}$$

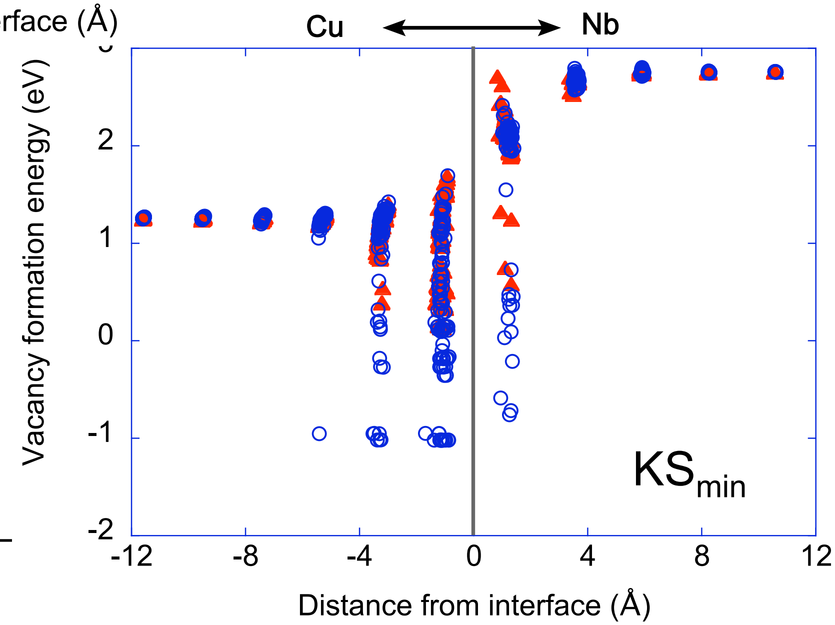
Results:

- Produced Cu interstitials are mostly loaded into the interface after cascade;
- Nb defects do not load significantly into the interface

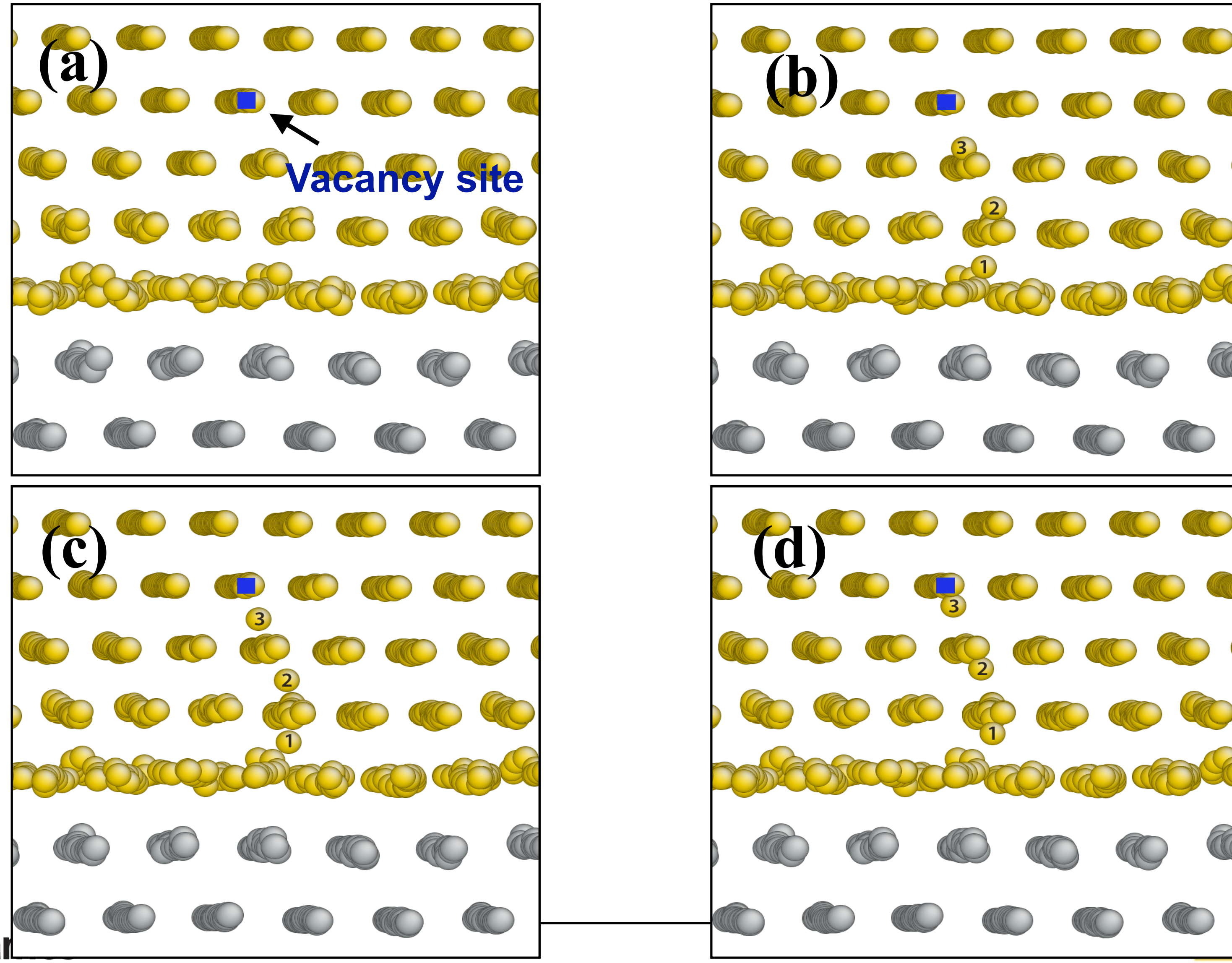
“Interstitial loading” effect on vacancy behavior via interstitial emission at KS interfaces



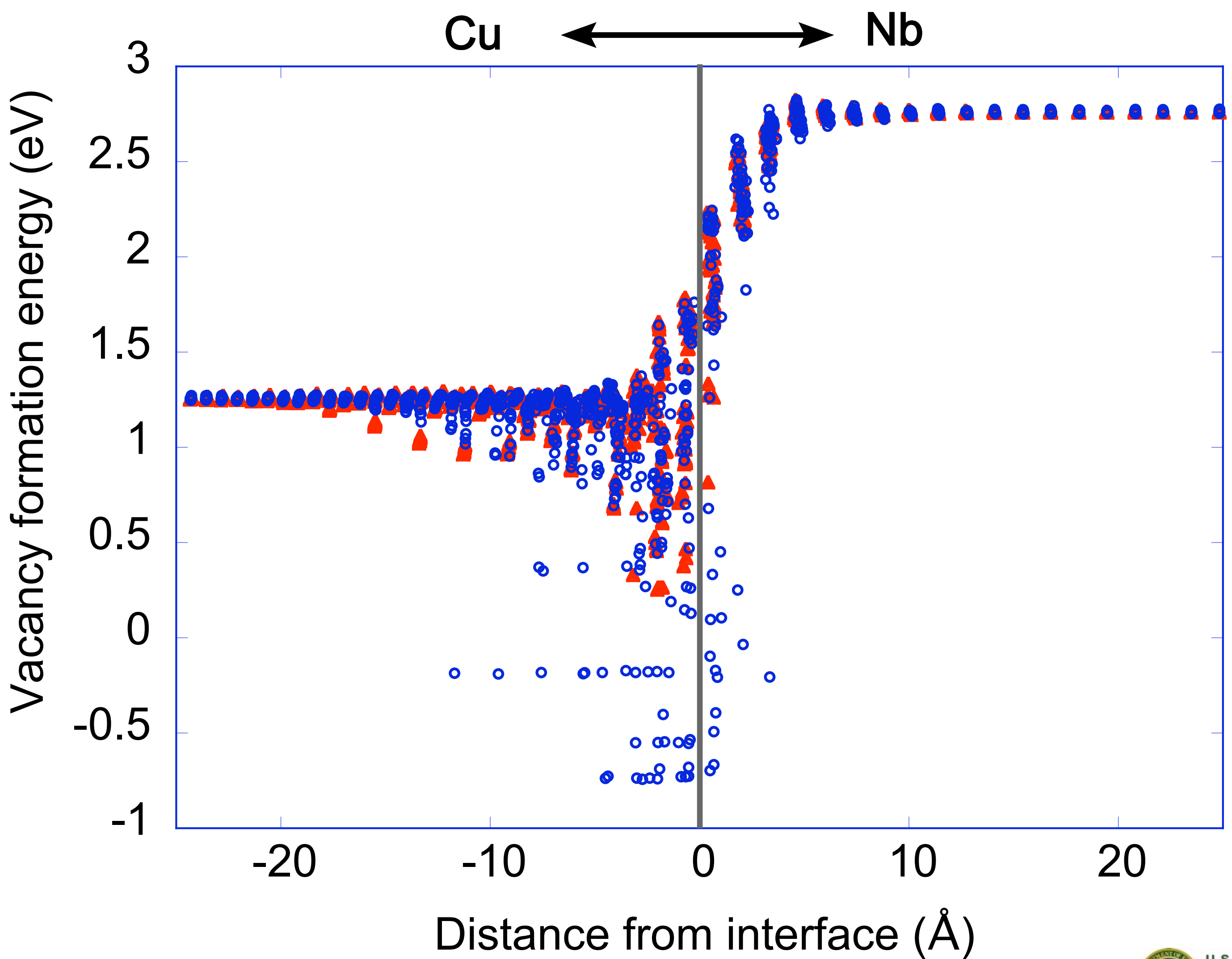




Atomistic processes in the observed interstitial emission and recombination events



"Interstitial loading" effect on vacancy behavior via interstitial emission at SPD interfaces

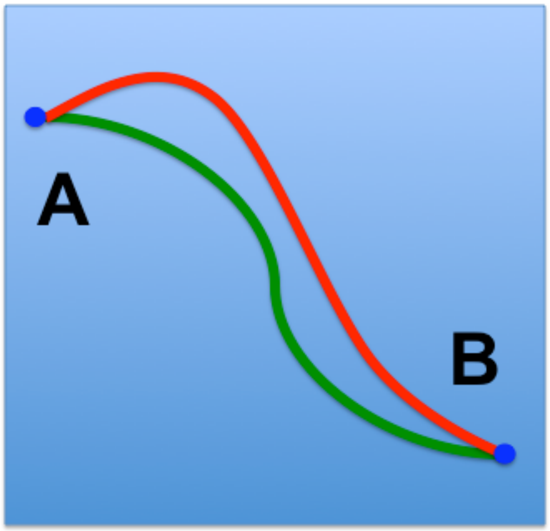


Loading of Nb interstitials at SPD Cu-Nb interface

• Nb interstitials like to aggolomerate to form relatively immobile clusters the Nb layer or are pushed into the Cu side, leading to intermixing developed at the interface.

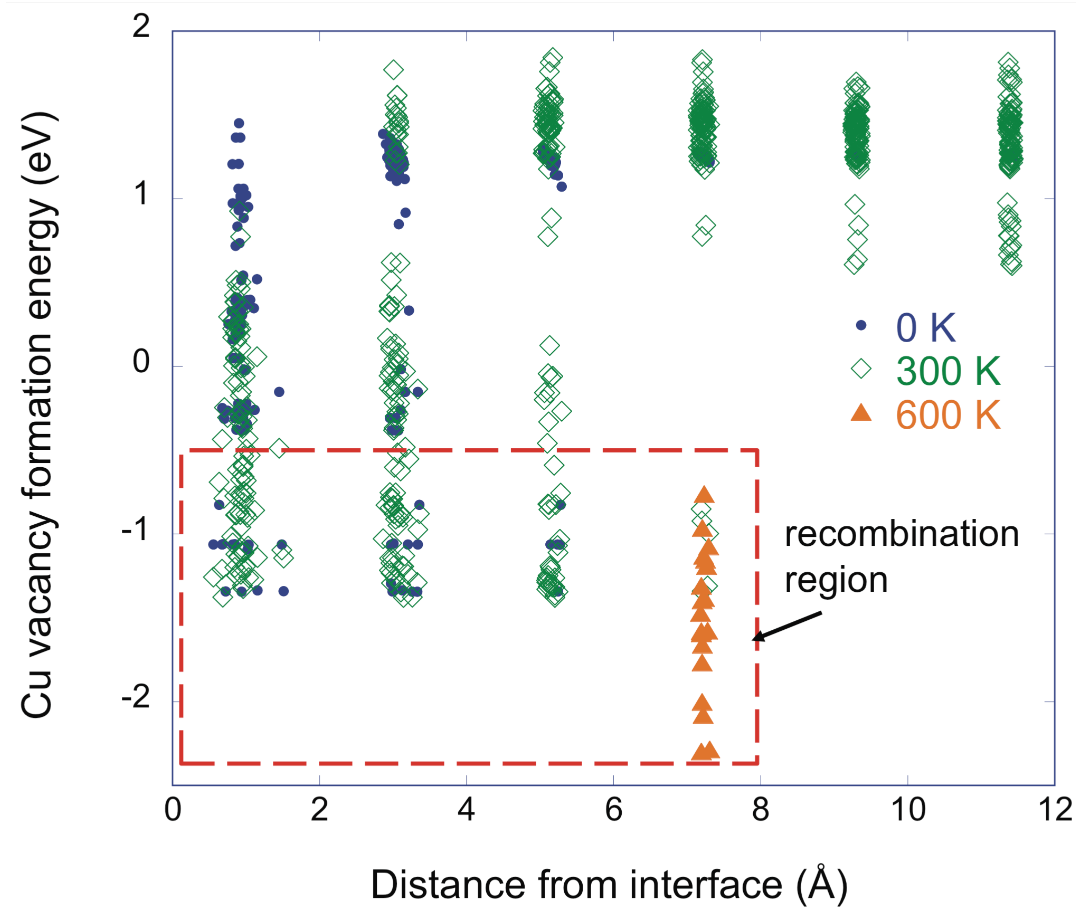
• Because of that, the Nb interstitial loading has no or negligible effect on vacancy-interstitial recombination on either Nb side or Cu side of the interface.

Low barriers for recombination events at "interstitial loaded" KS_1 interface



MD simulations
- 300 K, 10 ps
- 600 K, 100 ps

600 K result: only recombined events at selected sites shown.



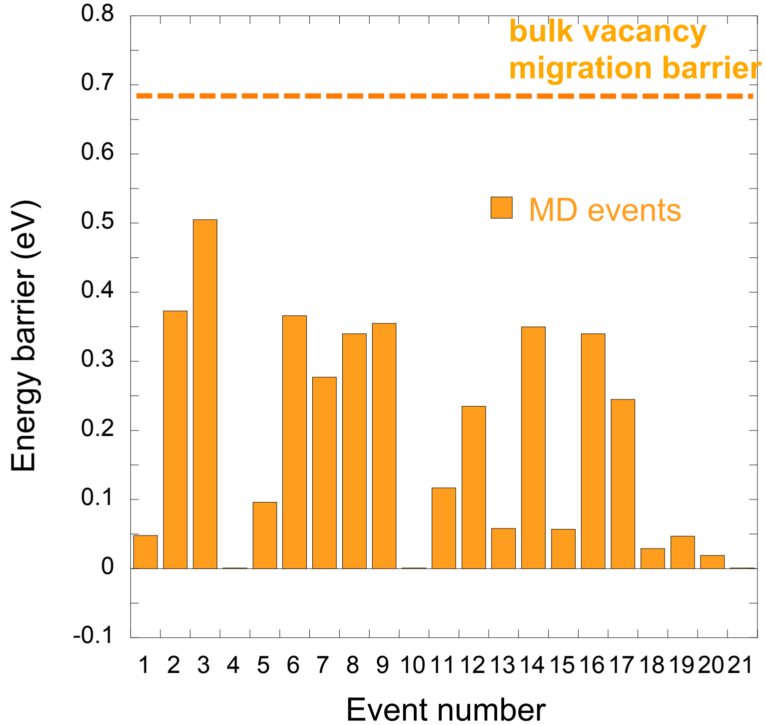
MD simulations at higher temperatures reveal "low barrier" recombination events

→ Larger interaction range

→ Higher density of recombination sites

Analysis of low barriers recombination events

- Nudged elastic band (NEB) method to calculate the transition states and energy barriers for all the events leading to recombination in 600 K MD simulations.



In these cases, interstitial emission occurs with a barrier much smaller than the bulk vacancy diffusion barrier

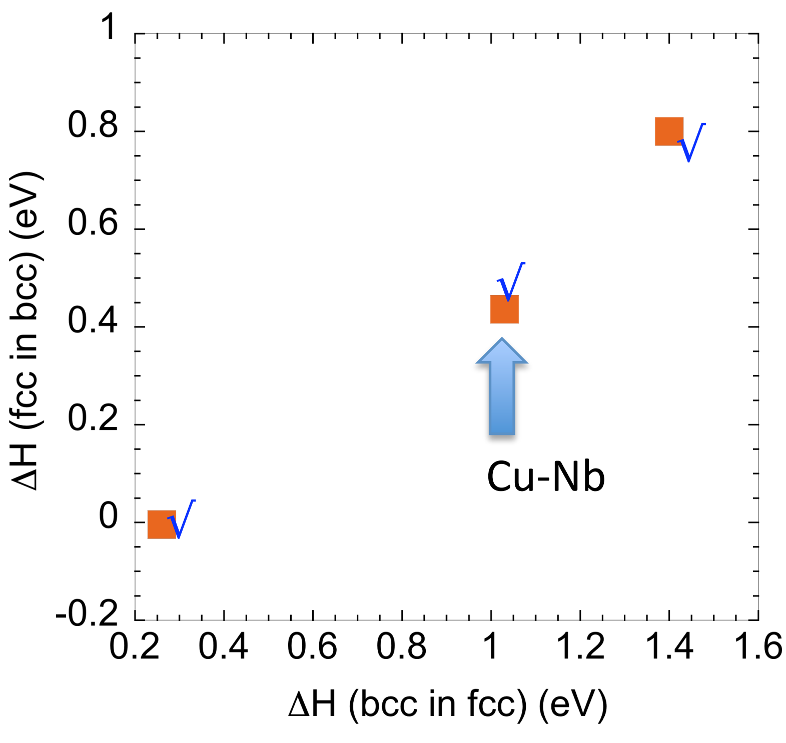
→ Interstitial emission much faster than vacancy diffusion

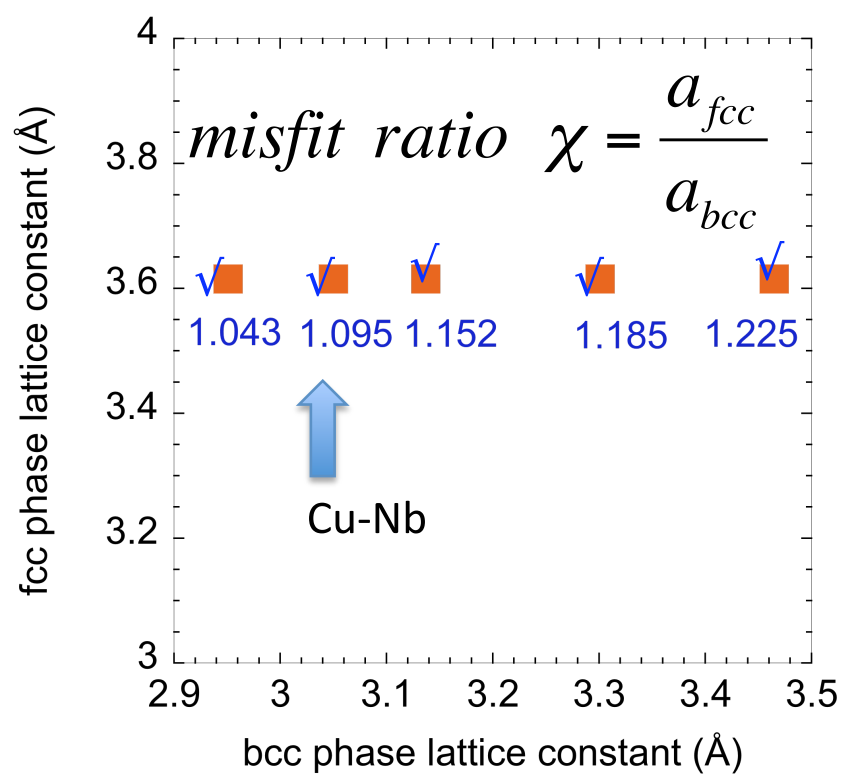
"Interstitial loading" effect at fcc-bcc KS interfaces is generic

Use a "tunable" potential approach to vary interface properties:

- Keep lattice misfit constant, systematically vary heats of mixing,
- Or, keep heats of mixing constant, systematically vary lattice misfit at interface.

√ = "unloading" effect observed at the Cu side of KS interfaces





Interstitial emission mechanism occurs at all interfaces studied.

Discussion on defect "identity"

- Regardless of the detailed structure of interfaces and the extent of point defect delocalization at interfaces, the interstitial loading-unloading effect exists and similar mechanism, i.e., the interstitial emission process occurs.

- This indicates that interstitials do not lose their identity when absorbed at these interfaces and thus retain the ability to interact with vacancies in the bulk region of the layer.

Summary

- MD studies of collision cascades in KS-type Cu-Nb interfaces are performed, revealing that Cu interstitials are preferentially loaded into the interface while on the bcc Nb side there is very little absorption of interstitials.

- The interstitial loading effect at two types of heterogeneous Cu-Nb interfaces, both KS-type and SPD {112}-type interfaces is studied:
  - The Cu interstitials are observed to spontaneously emit from both types of interfaces to annihilate vacancies in the nearby bulk.
  - MD and NEB studies are used to characterize low barrier emission processes.

- Our study also indicates that interstitials do not lose their identity when absorbed even at interfaces where they become significantly delocalized.